



Electron beam technology for biogas and biofertilizer generation at municipal resource recovery facilities

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Abstract. In the era of circular economies, municipal wastewater treatment plants (WWTPs) are viewed as resource recovery facilities. At the very minimum, the targeted resources are water, biogas, and phosphorus. However, municipal wastewater streams (sludge and effluent) need to be adequately treated to eliminate the potential for the transmission of microbial pathogens including protozoa, bacteria, and viruses. This paper presents the results from a study demonstrating the use of electron beam technology for sludge hygenization and enhanced methane (biogas) production using municipal wastewater samples. Cogeneration of heat for fertilizer drying and granulation and electricity for powering the electron beam system are also demonstrated.

Keywords: Biogas • Biofertilizer • Cogeneration • Electron beam • Ionizing radiation • Municipal wastewater plant sludge • Phosphorus • Resource recovery facility • Sludge hygenization

Introduction

Circular economy at wastewater treatment plants (WWTPs)

Urbanization is occurring rapidly around the world. More than half of the world's population now lives in urban areas. This has resulted in significant increases in population densities resulting in "megacities" [1]. These megacities are found in almost all major countries around the world. In 1950, there were only two cities in the world with a population greater than 10 million. Sorensen and Okata [1] estimate that by 2025, there will be approximately 27 cities around the world with more than 10 million people. In all, 22 out of these 27 megacities will be in the developing parts of the world [2]. It should be borne in mind that the exact definition of a megacity is debatable. Increasing urbanization calls for effective management of human wastes. It is abundantly clear that for these urban areas to be sustainable, public health must be protected. The conventional view has been that to protect public health, there needs to be proper collection, treatment, and disposal of municipal solid and liquid wastes. However, human waste streams are significant pools of water, energy substrates, and nutrients. Given the value of these resources, in today's era of circular economies, the

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concept of WWTPs has been replaced by the concept of these treatment plants as “resource recovery facilities” [3, 4]. The water from human waste streams can be recycled with appropriate treatment technologies for reuse purposes, the organic fraction within these waste streams can be harnessed to yield energy, and the phosphorus content of the wastes could be harvested for inorganic fertilizer use. According to the data of the Polish Central Statistical Office, 513 000 tons of dry mass (d.m.) municipal sewage sludge was used and stored in Poland, in 2012. The average content of phosphorus in them was at the level of 1.83%. Thus, the produced sludge contained 9400 tons of phosphorus. The agricultural use is regulated in the European Union (EU) by the Directive EU/2018/851 [5, 6].

Radiation technologies for sludge treatment

The use of ionizing radiation technology such as electron beam (e-beam) treatment for sludge hygienization is not new. In the United States, the EPA has already approved the use of ionizing radiation technology at 10 kGy as a process to further reduce pathogens (PFRP) to yield Class A [7] biosolids. A pilot-scale low energy e-beam wastewater treatment plant was operational in Florida in the early 1990s. Praveen *et al.* [8] have reported that e-beam irradiation technology is effective against a variety of microbial pathogens and fecal indicator organisms. The efficacy of e-beam technology at the pilot-scale level has already been demonstrated in Poland [9], Korea [10, 11], and other countries [12]. The role of water radiolysis on the sludge disintegration process and direct and indirect DNA damage for pathogen inactivation was discussed in more detail in [13, 14]. This paper presents the concept of combining electron beam sludge treatment technology with biogas production in an industrial plant that is equipped to generate electricity to power the accelerator. This technology, providing organic fertilizer, biogas, and electricity fits well into the circular economy concept [15]. The literature cited discussed pathogen deactivation and influence of radiation on the sludge physical parameters. The significance and novelty of the findings in our paper refer to the influence of radiation on sludge disintegration and the resultant increase in the rate of anaerobic fermentation.

Accelerator systems to be used for sludge treatment

The engineering and technology attributes of accelerators suitable for environmental applications have been previously reviewed by Zimek [16]. Although the beam power of accelerators has improved over the past decade, and there have been changes in the electronic elements of the control systems, no dramatic changes have occurred in their operating principles and design since then. With regard to environmental applications, the high-power transformer-based accelerators with beam energy range of 1–2 MeV may be preferred because of their high-

-beam power capabilities. Moreover, these units are of high-energy efficiency (plug to beam conversion) and the capital expenses are relatively modest. On the other hand, the major drawback is the low penetration of the electrons from such accelerators, which requires that special consideration needs to be paid to designing the beam and product handling system to facilitate the low energy electrons. The application of e-beam technology using accelerators as an environmental treatment technology was reviewed in Chmielewski [17] and Chmielewski & Han [18].

There were two key objectives in this study. One was to understand the influence of e-beam irradiation as a sludge pretreatment on biogas yield and the other was to evaluate whether e-beam treatment of municipal waste for hygienization could be harnessed to produce organic fertilizers.

Materials and methods

Sludge samples used in investigations

The experiments focusing on the influence of e-beam treatment on biogas production efficiency were at a wastewater treatment plant located in south east Poland (SEP WWTP). The SEP WWTP treats primarily wastewater flows from agricultural industries manufacturing jams, pickles, and such fruit products. Only a small portion of the wastewater flow comes from domestic sources. The scheme of this plant is presented in Fig. 1.

Electron beam accelerator units

The e-beam trials were performed using the ILU-6 electron accelerator and the 10/10 Elektronika accelerators [19]. The samples were contained within custom-designed cassettes. The aluminium cassette (measuring 400 mm × 100 mm) had a lid with a titanium foil-(50 µm) covered window. The cassette could hold a total of 100 ml volume of the test samples creating a 2.5-mm thick sludge sample within the cassette. Similarly, when the 10/10 Elektronika accelerator was used, the sludge samples were contained within custom-designed cassettes that could hold 1500 mL of sludge material. The cassettes were double-welded foil packages. For dosimetry, PVC strips, CTA strips (cellulose triacetate), or Harwell 3042 dosimeters were used.

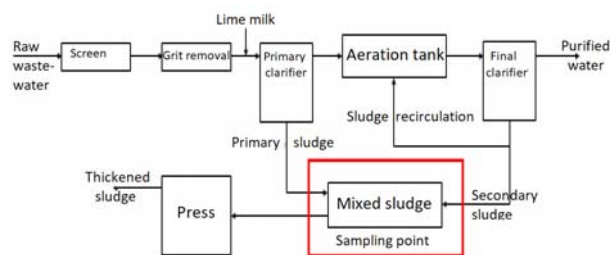


Fig. 1. Schematic of the SEP WWTP and the sampling location (marked in red).



Fig. 2. DIN 38414/8 eudiometers used for small-scale (400 mL) experiments including water bath, Testo 622 instrument for temperature and atmospheric pressure measurements table specially designed for this purpose.

Methane generation studies

The methane generation studies were carried out in 400-mL bioreactors (under mesophilic conditions) connected with eudiometer tubes compliant with DIN38414/8 standard (Fig. 2) (Behr Labor-Technik GmbH, Düsseldorf, Germany) (Table 1). The sludge samples used in these studies comprised of the non e-beam treated control samples and sludge samples exposed to 1 kGy, 2 kGy, and 3 kGy e-beam target doses. Aliquots of the biogas digester digestate from the SEP WWTP were used as the inoculum. The inoculum to biomass ratio in the experimental bioreactor mixture was 20/80 in all the experiments. Sodium bicarbonate was used to adjust the pH of the bioreactor after the addition of the inoculum and the samples. Three replicate 400-mL bioreactors were run concurrently for each dose and for the 0 kGy control. Due to the limitations of the available bioreactors, methane formation studies for the different e-beam doses were performed sequentially.

The bioreactors were operated at approximately 38°C using Labo Play W620 waterbath (Laboplay, Bytom, Poland) for a total duration of 21 days. The content of the bioreactors was stirred manually every 24 h just before measuring the biogas output volume. The initial and final pH within the bioreactor were measured using the commercially available Elmetron CX-105 multifunction meter (Elmetron, Zabrze, Poland) combined with Elmetron GPX-105s head (designed to work with sludges and pulps). A Testo 622 instrument (Testo, Titisee-Neustadt, Germany) was used to measure the atmospheric pressure and the ambient temperature. The chemical oxygen demand (COD) in the liquid phase (soluble chemical oxygen demand – SCOD) was measured before and after e-beam irradiation as well as before and after the methane fermentation process. To measure SCOD, 15 mL samples were centrifuged at 5100 rpm for 30 min using MPW-54 (MPW Med. Instruments, Warsaw, Poland) centrifuge. The supernatant was filtered using (0.45 µm) filters (VWR, Pennsylvania, USA) and analysed using Macherey–Nagel photo-

Table 1. Bioreactor conditions to monitor methane generation

Source of sludge	SEP WWTP
Source of inoculum	Digestate from SEP WWTP biogas digester
Inoculum: Substrate ratio	20%:80%
Bioreactor pH	pH 7.1–7.3
Bioreactor volume	400 ml
Residence time	21 days
Study temperature	38°C
Mixing conditions	Manually, once every 24 h

metric tests and Macherey–Nagel Nanocolr Vis II photometer (Macherey–Nagel, Düren, Germany). The total suspended solids (TS) content (d. m.) in bioreactor mixture was measured before and after the fermentation by initial drying (103°C, 48 h). The organic mass content in dried bioreactor mixture (VS) was measured by loss during combustion at 530°C using PSK-31 furnace (Elterma, Świebodzin, Poland).

Statistical analysis

The statistical analysis that was performed during these studies was the student *t*-test using StatSoft Statistica software. Microsoft Excel (2019) was used for calculating the mean *v* and standard deviation values. Graphs were prepared using the same software.

Results

Methane generation from SEP WWTP

This sludge at the SEP WWTP originates primarily from agro-industries; hence, the pH was relatively low (6.8–6.9). For these experiments, the sludge after biological treatment and settling at the SEP WWTP was used as the starting material. The samples were placed in the bioreactors and the methane generation was monitored for 21 days under mesophilic conditions (Table 1). Three replicate bioreactor studies were performed for each treatment. Both the e-beam treated and the untreated (control) sludges were placed in separate bioreactor vessels to monitor methane generation (Figs. 3A–C).

There was a difference in the methane concentrations and the kinetics of methane generation from the sludge obtained from the SEP WWTP, irrespective of whether it was e-beam treated or not (Figs. 3A–C). The untreated control samples showed significantly different concentrations of methane at the end of 21 days. This suggests that there is significant difference in the substrate concentration in the sludge samples obtained on different days. This is reflected in the SCOD levels in the sludge samples obtained on the different days (Table 2). It is necessary to point out that these data represent realistic incoming wastewater conditions since the agricultural industry wastewater treated in the plant probably changes depending on the fruits/vegetables processed and the yield of production, which change

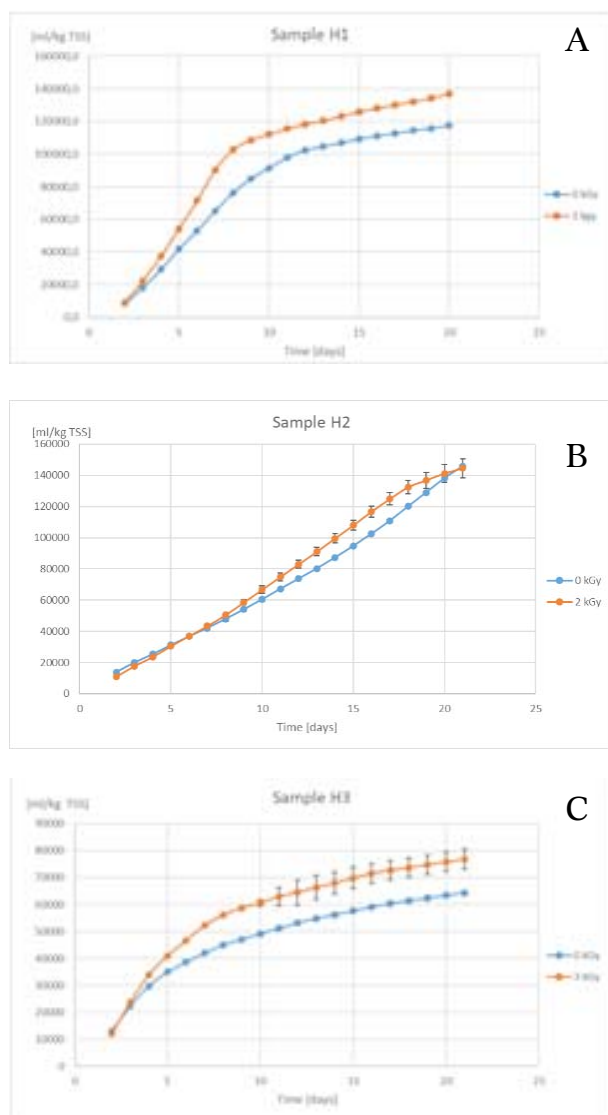


Fig. 3. Methane generation over 21 days mesophilic digestion of wastewater treatment plant sludge pretreated at 1 kGy (A), 2 kGy (B) and 3 kGy (C) e-beam doses and data for references samples not irradiated (0 kGy). H1–H3 represent independent experiments performed on separate days using different sludge samples.

the ratio prevailing between the rates of municipal/agricultural wastewater streams. Nevertheless, the effect of e-beam treatment on the sludge samples in terms of biogas yield is evident. It is well known that e-beam treatment does not breakdown to microbial cells. Therefore, this implies that the effect of

e-beam treatment even at low e-beam doses such as 2 kGy and 3 kGy is capable of changing the sludge characteristics, resulting in enhanced biogas yields. Other investigators [20, 21] have also reported similar results that e-beam irradiation does change the sludge characteristics. The biogas yield which normally takes approximately 21 days was achieved in 11–14 days (H1 and H3) at the same process conditions. Degradation of the biomass structure and observed higher yield of biomass production from an existing plant at which e-beam system has been applied (retrofit) or allows in the case of a newly built plant, equipped in such a system, to construct smaller volume installation with the same planned production of methane. The lack of significant differences between the control and e-beam treated samples in study H2 reflects the effect of incoming wastewater quality resulting from the excess sludge parameters on the biogas yield for the irradiated sample. However, increase for the biogas production (irradiated sample) has been observed till day 18; the biogas yield was higher for H2 in comparison to H1 and H3 and this was probably due to the fact that nutrient content (leading to change in carbon to nitrogen (C/N) ratio) was already consumed by methanogens (day 21). As mentioned earlier, the varying wastewater quality on the three sampling days can be detected by the biogas yield in the control samples. The varying results observed in the wastewater samples from the three separate sampling days imply that the quality of the wastewater for biogas yield has to meet certain specifications. The results from the H1 and H3 studies imply that biogas production from agro-industrial wastewater is possible and subsequent cogeneration of heat for electrical power is a possibility.

Many methods of disintegration of sewage sludge have been developed, and all of them are based on the addition of energy input into biomass substrate processing. The energy required to be supplied as input can be delivered by different methods; these are mechanical, physical, chemical, biological, and hybrid [22]. Electron beam processing is based on physical energy transfer (kGy value gives an energy input in kJ per kg of irradiated matter) followed by chemical processes in which water radiolysis products play a very important role. This mechanism is described in a recently published work [13]. The process of disintegration of excess sludge, being a feed to anaerobic digesters, results in the higher production of biogas, and a lower concentration

Table 2. Cumulative SCOD (mg/L) and methane volumes generated (ml/kg TS) as a function of e-beam dose

Experiment	Dose (kGy)	Volume of biogas produced after 21 days (ml/kg TS)	<i>t</i> -test $\alpha = 0.05$	SCOD (mgO ₂ /L) before the fermentation	<i>t</i> -test $\alpha = 0.05$
H1	0	118.842 ± 721	$t = -27.0145$	–	–
	1	138.602 ± 1630	$p = 0.0014$	–	–
H2	0	145.822 ± 1003	$t = 0.4654$	4993 ± 24	$t = -18.0121$
	2	144.447 ± 6074	$p = 0.6874$	5754 ± 59	$p = 0.0031$
H3	0	64.374 ± 910	$t = -7.2157$	318 ± 0	$t = -664.0$
	3	76.844 ± 3647	$p = 0.0187$	1425 ± 3	$p = 0.000002$

of organic dry mass in digestate, improving its susceptibility to the dewatering processes, which is demonstrated by the higher SCOD values, and this means that the concentration of the nutrients is in both the hydrolysis and fermentation steps. The authors in this work focus only on the biogas production; visible differences in the ratio of CH₄ to CO₂, for both unirradiated and irradiated sludge, were not noticed. However, measurement of the H₂ concentration in biogas planned for next experiments may give an answer to how e-beam affects the overall gas composition (CH₄, H₂, CO₂). The higher dose effects on the process will be tested, as well.

Other advantages of the process that are related to the phenomena reported in the previous studies [14] have demonstrated the destruction of parasites and their eggs resulting in sludge disinfection. Additional studies are, however, needed to determine the dose required to eliminate bacterial and viral pathogens so that the sludge can be used for land application with minimal restrictions. The US Environmental Protection Agency (US EPA) has already established a minimum dose of 10 kGy as a PFRP. It will be interesting to understand the biogas yield from agro-industrial wastewater when 10 kGy is used for sludge pretreatment. Park *et al.* [20] have reported that biogas yields improve by as much as 22% even at 7 kGy.

Municipal WWTPs around the world already operate biogas recovery equipment. Therefore, appropriately sized e-beam accelerator equipment and

material handling systems can be installed within these plants. The optimal solids content to achieve enhanced biogas recovery needs further studies. Agronomic studies are also needed to demonstrate the nutrients that can be recovered from municipal sludges that have been microbially decontaminated (by e-beam treatment) and the biogas can be recovered by anaerobic digestion. The advantage of having the digester downstream of the e-beam treatment step is that the biosolids from the digester will be considered “stabilized” (per the USEPA standards) for vector attraction. One can envision the augmentation of biogas generation by incorporating additional biomass, including green waste such as grass silage and landscaping wastes. A preliminary economic analysis in terms of cost savings associated the use of e-beam technology for sludge hygienization primarily compared with use of e-beam technology for sludge hygienization and biogas cogeneration, as shown in Table 3.

The above-described economic analysis is based on a small municipal wastewater treatment plant serving a population of approximately 10500 individuals. Combining low capital expense e-beam accelerator technology (100 kW, 2 MeV) with anaerobic digestion opens up several possibilities for converting a traditional wastewater treatment plant into a resource recovery facility. Resource recovery facilities such as these provide a financially and technologically sustainable operation to generate

Table 3. Preliminary economic analysis of incorporating e-beam technology for sludge hygienization solely compared with incorporating e-beam technology for sludge hygienization and biogas cogeneration

Wastewater treatment plant	
(Throughput: ~250 000 m ³ annually. Sludge output ~1500 tons dry mass annually)	
I. E-beam technology for sludge hygienization	II. E-beam technology for sludge hygienization and biogas cogeneration
<i>Accelerator specification</i>	
100 kW, 2 MeV	100 kW, 2 MeV + biogas generation
Cost savings	
<i>Savings from avoiding sludge disposal costs</i>	
1500 tons @ 100 euros = 150 000 euros	1500 tons @ 100 euros = 150 000 euros
<i>Potential revenue from biosolid-based fertilizer sales</i>	
1500 tons @ 94.5 euros = 141 750 euros	1277.5 tons @ 94.5 euros = 120 723.75 euros
	Biogas production (1 022 000 m ³ annually) Converted in co-generator in electricity and heat Generator power 350 kW Equivalent of electricity production: 350 kW × 8000 h × 0.13 euros/kWh = 354 000 euros
E-beam technology-associated operating costs	
<i>Electricity consumption</i>	
130 kW e-beam accelerator 70 kW wastewater plant equipment 10 kW heat generation	130 kW e-beam accelerator 70 kW wastewater plant equipment
<i>Total cost</i>	
210 kW × 8000 h × 0.13 euros/kwh = 218 400 euros	200 kW × 8000 h × 0.13 euros/kwh = 208 000 euros 1055 tons grass silage (annually) = 1 055 tons × 10 euros/ton = 10 055 euros
Net income and savings	
73 350 euros annually	271 668.75 euros annually



both biogas and fertilizers for usage by the surrounding communities. Importantly, the incorporation of e-beam technology into a wastewater treatment plant will result in significant cost savings.

Conclusions

1. Sludge irradiation can increase the biogas yield during downstream anaerobic fermentation (digester) process.
2. The breakdown of the soluble and suspended organic matter possibly leads to SCOD increase and the availability of additional nutrients for digester performance.
3. The yield of biogas production obtained in 11–14 days was comparable to the biogas obtained in 21 days in untreated samples at the same fermentation process conditions. The ability to reduce digester residence times without reducing biogas production has major economic and process implications. Optimization studies can further improve biogas production efficiency as well as reduce digester residence times.
4. This study has demonstrated that small WWTPs serving industrial or residential waste streams could be retrofitted with appropriately sized e-beam equipment to convert them into true resource recovery facilities.
5. Agronomic studies are needed to demonstrate the recovery of plant nutrients from such e-beam-treated sludge samples.

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